



THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant(s) : Bi Le-Khac et al.
Appl. No. : 09/760,931
Filed : January 16, 2001
Title : PROCESS FOR PREPARING COMB-
BRANCHED POLYMERS
Group Art Unit : 1713
Examiner : Tatyana Zalukaeva
Docket No. : 01-2532B

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1/9/03

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Honorable Commissioner for Patents
Washington, DC 20231

APPEAL BRIEF UNDER 37 C.F.R. § 1.192(d)

I. REAL PARTY IN INTEREST

The real party in interest is Equistar Chemicals, LP.

II. RELATED APPEALS AND INTERFERENCES

There are no other appeals or interferences known to Applicants, their representatives, or their assignee that will directly affect or be directly affected by or have a bearing on the Board's decision in this appeal.

III. STATUS OF CLAIMS

Claims 1-5, 7, 9, 10, 12 and 13 are on appeal.

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IV. GROUPING OF CLAIMS

The claims on appeal shall stand or fall together.

V. STATUS OF AMENDMENTS

Claims on appeal have not been amended after the Final Office Action.

VI. SUMMARY OF THE INVENTION

The invention is a continuous process for making comb-branched copolymers of a polyether macromonomer and an acrylic comonomer. The comb-branched copolymers are used as water reducing agents in cement. The process comprises (a) forming a monomer stream containing an acrylic monomer, a polyether macromonomer, and a chain transfer agent; an initiator stream; and, optionally, a chain transfer agent stream; ~~(b) polymerizing the~~ streams; and (c) continuously withdrawing a polymer stream. See claim 1.

VII. QUESTION PRESENTED FOR REVIEW

Whether *Nagano* (U.S. Pat. No. 5,834,576) anticipates the Applicants' claimed process in which the use of chain transfer agent is an essential element, while *Nagano* is silent in this aspect.

VIII. ARGUMENTS

"A claim is anticipated only if each and every element as set forth in the claim is found." *MPEP* § 2131; see also *Verdegaal Bros. v. Union Oil Co. of California*, 814 F.2d 628, 631, 2 USPQ2d 1051, 1053 (Fed. Cir. 1987). Applicants respectfully contend that the Examiner has erred in rejecting the claims as being anticipated by *Nagano* while she recognizes that *Nagano* is silent about the use of chain transfer agent, which is an essential element of the claims. Applicants also contend that the Examiner has erred in rejecting Applicants' claims based on *Nagano*'s teachings of oxygen and phenothiazine because such teachings are irrelevant to the claims and because oxygen and phenothiazine are well known inhibitors but not chain transfer agents.

A. NAGANO CANNOT ANTICIPATE APPLICANTS' CLAIMS BECAUSE NAGANO IS SILENT ABOUT THE USE OF CHAIN TRANSFER AGENTS.

Nagano cannot anticipate the Applicants' invention because it fails to teach the use of chain transfer agents in the claimed process. Applicants' process of the invention is a free radical copolymerization in which a polyether macromonomer is copolymerized with an acrylic comonomer in the presence of a free radical initiator. The process produces a comb-branched copolymer. The process comprises three steps: (a) forming a monomer stream containing an acrylic monomer, a polyether macromonomer, and a chain transfer agent; an initiator stream; and, optionally, a chain transfer agent stream; (b) polymerizing

the streams; and (c) continuously withdrawing a polymer stream. See, e.g., *claim 1*.

The use of chain transfer agent is an essential element of the claims. Applicants also clearly teach in the specification what kinds of chain transfer agents would be suitable for the use in the process. In the DETAILED DESCRIPTION OF THE INVENTION, Applicants teach that suitable chain transfer agent includes alkyl amines, alkyl sulfides, alkyl disulfides, carbon tetrahalides, allyl ethers, and mercaptans. See *page 5, lines 25-28 of the application*. Applicants further teach that preferred chain transfer agents are mercaptans. *Id.* Further, in all examples, Applicants use mercaptopropionic acid as a chain transfer agent. See *Examples on pages 8-11 of the application*.

~~In contrast, Nagano in nowhere teaches the use of any chain transfer agents. Nagano's teachings can be divided into two parts:~~

- (I) The preparation of polyether macromonomers, including col. 2, line 43, to col. 21, line 37, col. 37, line 9, to col. 40, and Examples 1, 6, 8, 10, 13-28.
- (II) Free radical polymerization of polyether macromonomers, including col. 21, line 38, to col. 37, line 7, and Examples 2-4, 7, 9, 11, 12, 21, 27, and 28.

Applicants note that only teachings of Group (II) are relevant to Applicants' claims. The teachings of Group (I) are irrelevant because they are related only to the preparation of polyether macromonomers, which is not a part of the

claims. Nevertheless, *Nagano* has no teachings of using any chain transfer agents in either Group (I) or (II).

In fact, the Examiner has admitted that "***Nagano* is silent about the use of chain transfer agents.**" See page 4 of the Office Action dated February 6, 2002. Given that the Examiner's only rejection to the claims is the anticipation rejection based on *Nagano* and that the Examiner recognizes that an essential element of the claims is missing from *Nagano*, the Examiner should have allowed the claims according to MPEP § 2131. However, the Examiner has failed to do so. Thus, Applicants respectfully ask the Honorable Board to reverse the Examiner's rejection.

**B. THE EXAMINER HAS ERRED IN REJECTING THE CLAIMS BASED ON
NAGANO'S TEACHINGS OF OXYGEN OR PHENOTHIAZINE
BECAUSE THE TEACHINGS ARE IRRELEVANT TO THE CLAIMS.**

The Examiner reverses her position that *Nagano* is silent about the use of chain transfer agent and gives Applicants' claims final rejection based on her new reading of *Nagano*. The Examiner asserts in the Final Office Action:

"With regard to using a chain transfer agent in a polymerization step, *Nagano* teaches that it is desirable to add molecular oxygen or air in order to restrain polymerization of acrylic ester and cyclic ether compound (column 12, lines 24-29), in other words to regulate the molecular weight of the chain. Further *Nagano* teaches that inhibitors of polymerization or combination of those, such as phenothiazine are used to such inhibitors in combination with molecular oxygen can be used.

(Column 12, lines 30-44)." (Emphasis used by the Examiner). See the last paragraph on page 3 of the Final Office Action dated August 19, 2002.

As discussed above, *Nagano*'s teachings from col. 12, lines 24 to 29, and from col. 12, lines 30 to 44, cited by the Examiner here, fall within Group (I). Group (I) only teaches the preparation of polyether macromonomers by a ring-opening polymerization. These teachings are irrelevant to Applicants' claims, which define a free radical copolymerization of a polyether macromonomer with an acrylic comonomer. Thus, the Examiner has clearly erred in relying on Group (I) of *Nagano* to reject the claims because the preparation of polyether macromonomer taught in Group (I) is not an "element as set forth in the claim."

See MPEP § 2131. Thus, the Examiner's rejection of the claims is improper and should be reversed.

C. EVEN IF THE EXAMINER COULD USE NAGANO'S GROUP (I), SHE HAS STILL ERRED BECAUSE OXYGEN AND PHENOTHIAZINE ARE INHIBITORS BUT NOT CHAIN TRANSFER AGENTS.

Contrary to the Examiner's assertion, *Nagano* teaches the use of oxygen and phenothiazine as inhibitors rather than as chain transfer agents. *Nagano*, in col. 12, lines 24-29, reads:

"Therefore, when reacting the acrylic ester with the cyclic ether compound, it is desirable to add a polymerization inhibitor or molecular oxygen to the system of reaction in order to restrain the polymerization of

the acrylic ester, the cyclic ether and the acrylic ester compound.”

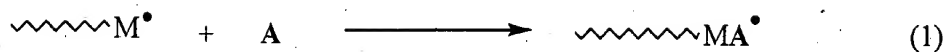
(Emphasis added).

In col. 12, lines 30-44, *Nagano* reads:

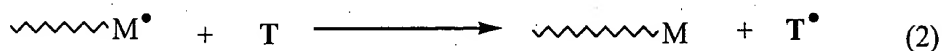
“The polymerization inhibitor is not particularly limited. Examples are hydroquinone, hydroquinone monoethyl ether, p-benzoquinone, t-butyl catechol, and phenothiazine. . . . In order to sufficiently restrain the polymerization, it is desirable to use the polymerization inhibitor and the molecular oxygen together.” (Emphasis added).

These languages clearly indicate that *Nagano* uses oxygen and phenothiazine as inhibitors rather than chain transfer agents. To reach a conclusion that oxygen and phenothiazine can be chain transfer agents, the Examiner cites three “extra” references: *Chamberlain* (U.S. Pat. No. 5,605,991), *Hoxmeier* (U.S. Pat. No. 5,773,521), and *Watson* (U.S. Pat. No. 4,466,904). She urges that a combination of *Nagano* with any of the three extra references can establish that oxygen and phenothiazine are chain transfer agents. See page 3 of the Office Action dated August 19, 2002. Applicants respectfully contend that the Examiner’s conclusion is wrong both legally and scientifically.

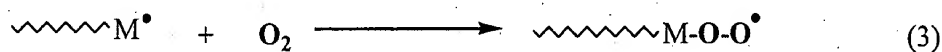
First, the Examiner’s conclusion is scientifically wrong. Inhibitors and chain transfer agents are terms of polymer science. They differ materially from each other. An inhibitor adds to a propagating radical chain to form a new radical that is inactive; it stops the propagation but does not form a new propagating chain (see scheme (1)). Therefore, inhibitors can completely stop a polymerization.



In contrast, a chain transfer agent terminates a propagating chain and forms a new, smaller radical that can grow a new chain (see scheme 2). Thus, a chain transfer agent is used to lower the molecular weight of the resulting polymer but not to stop the polymerization. See *Paul J. Flory, Principles of Polymer Chemistry, 1990, page 161.*



As *Nagano* clearly states, phenothiazine is an inhibitor. It is also well established that oxygen is an inhibitor. Oxygen reacts with a propagating polymer chain to form peroxide radical (see scheme (3)). See *Flory at page 168.* The peroxide radical has rather low reactivity and thus it stops the polymerization. *Id.* Thus, *Nagano's* teachings of using oxygen and phenothiazine as inhibitors are clear and consistent with the generally accepted scientific principles.



Second, because *Nagano's* use of oxygen and phenothiazine as inhibitors is clear, the Examiner's use of "extra" references in the anticipation rejection is improper under MPEP §2131. MPEP §2131.01 II provides that "extrinsic evidence may be used to explain but not expand the meaning of terms and

phrases used in the reference relied upon as anticipatory of the claimed subject matter.” (Emphasis added.) *In re Baxter Travenol Labs.*, 952 F.2d 388, 21 USPQ2d 1281 (Fed. Cir. 1991). Here, the Examiner not only expands the meaning of terms used in *Nagano* but also changes the meaning of terms that are clearly stated in the reference. Thus, the Examiner’s use of *Chamberlain*, *Hoxmeier*, and *Watson* is improper and her rejection of the claims based on the extra references should be reversed.

Finally, even if the Examiner could properly use any extra references, *Chamberlain*, *Hoxmeier*, or *Watson* cannot help the Examiner to establish that oxygen and phenothiazine are chain transfer agents in a free radical polymerization process.

Both *Chamberlain* and *Hoxmeier* teach only anionic polymerization.

Anionic polymerization differs from free radical polymerization in mechanism. They have different chain initiations, chain propagations, chain transfers, and chain terminations. A free radical initiator cannot start an anionic polymerization. Similarly, a free radical chain transfer agent is not likely to be a chain transfer agent in an anionic polymerization. Given that *Chamberlain* and *Hoxmeier* use oxygen as a termination agent or *chain transfer agent* in anionic polymerization, this does not establish that oxygen is a chain transfer agent in a free radical polymerization. In fact, oxygen is very well known to be a powerful chain transfer agent in a free radical polymerization as discussed above.

Similarly, *Watson* does not help the Examiner to establish that phenothiazine can be a chain transfer in Applicants’ claimed process. *Watson*

teaches using phenothiazine as an inhibitor in the distillation of aromatic monomers. That phenothiazine is an inhibitor is clearly stated by *Nagano*. Thus, *Watson* adds nothing to *Nagano*.

In the Advisory Action, the Examiner argues: "Compounds of identical chemical composition cannot have mutually exclusive properties." This argument is fundamentally flawed because a chemical compound may, and often does, have different chemical properties in different reactions.

In conclusion, *Nagano* cannot anticipate Applicants' claims because (1) *Nagano* does not teach the use of any chain transfer agents; (2) *Nagano*'s teachings of oxygen and phenothiazine are irrelevant to the claims; and (3) oxygen and phenothiazine are inhibitors but not chain transfer agents. Therefore, Applicants respectfully ask the Honorable Board to reverse the

Examiner's rejection and to allow claims 1-5, 7, 9, 10, 12, and 13.

Respectfully submitted,
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12/31/2002

Appendix

Claims on Appeal

1. A continuous process which comprises:
 - (a) forming a monomer stream that contains an acrylic monomer, a polyether macromonomer selected from the group consisting of poly(propylene glycol) acrylate, poly(propylene glycol) methacrylate, poly(ethylene glycol) acrylate, poly(ethylene glycol) methacrylate, acrylates and methacrylates of an oxyethylene and oxypropylene block and random copolymer, and mixtures thereof, and a chain transfer agent; and an initiator stream that contains a free radical initiator; and, optionally, a chain transfer agent stream;
 - (b) polymerizing the streams in a reactor at a temperature within the range of about -20°C to about 150°C ; and
 - (c) withdrawing a polymer stream from the reactor.

2. The process of claim 1 wherein the polymerization temperature is within the range of about 20°C to about 90°C .
3. The process of claim 1 wherein the polymerization temperature is within the range of about 40°C to about 60°C .
4. The process of claim 1 wherein the monomer stream further contains a solvent.
5. The process of claim 1 wherein the initiator stream further contains a solvent.
7. The process of claim 1 wherein the initiator stream further contains a chain transfer agent.

9. The process of claim 1 wherein the acrylic monomer is selected from the group consisting of acrylic acid, methacrylic acid, sodium acrylate, sodium methacrylate, ammonium acrylate, ammonium methacrylate, potassium acrylate, potassium methacrylate, and mixtures thereof.

10. The process of claim 1 wherein the initiator is a persulfate.

12. The process of claim 4 wherein the solvent is selected from the group consisting of water, methyl alcohol, ethyl alcohol, butyl alcohol, and isopropyl alcohol, and mixtures thereof.

13. The process of claim 5 wherein the solvent is selected from the group consisting of water, methyl alcohol, ethyl alcohol, butyl alcohol, and isopropyl alcohol.

PRINCIPLES OF
POLYMER CHEMISTRY

* PAUL J. FLORY *

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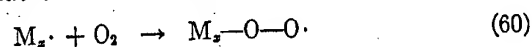
ing polymer which is not soluble in the reaction medium.⁸⁶ It is more closely related to the phenomenon of emulsion polymerization to be discussed in the following chapter than to autoacceleration in homogeneous polymerizing systems, however. The one feature in common is the decrease in (apparent) k_t caused by the reaction environment. However, the environmental factors obviously differ.

Absolute values of the rate constants for other reactions occurring in polymerizations and copolymerizations are readily obtained once k_p and k_t have been determined. Rate constants for chain transfer between growing radical and various species may be assigned absolute values simply by multiplying the appropriate transfer constant C (see Tables XIII, XIV and XV) by k_p , as previously indicated. By comparison of k_{tr} for different radicals with the same transfer agent, a direct measure of radical reactivity may be obtained. The chain transfer studies conducted up to the present time have been concerned with a variety of transfer agents and relatively few radicals; hence they are of limited use for this purpose. As will appear in Chapter V, k_p and k_t are essential for the assignment of absolute values to the rate constants for the cross propagation and termination processes which assume importance in copolymerizations.

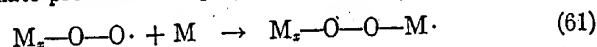
4. INHIBITION AND RETARDATION OF POLYMERIZATION

If there is added to the monomer a substance which reacts with chain radicals to yield either nonradical products or radicals of such low reactivity as to be incapable of adding monomer, the normal growth of polymer chains will be suppressed. The substance is designated an *inhibitor* if it is so effective as to reduce the polymerization rate substantially to zero. If, however, it acts less efficiently so that the rate and the degree of polymerization are reduced without total suppression of the polymerization, the substance is referred to as a *retarder*. The difference is, of course, one of degree. Results of Schulz⁸⁷ illustrating inhibition and retardation in the thermal polymerization of styrene are shown in Fig. 21. In the presence of 0.10 percent of benzoquinone, curve II, no measurable polymerization occurs throughout the *induction period*, during which the quinone is consumed by the thermally generated radicals. Thereafter the polymerization proceeds according to its normal course in the absence of inhibitor (curve I). A greater quantity of nitrobenzene (curve III) depresses the rate but without introducing an induction period. Nitrosobenzene (curve IV) causes inhibition, but following the induction period the rate remains lower than that for the pure monomer. The product resulting from the action of chain radicals on nitrosobenzene evidently is a retarder.

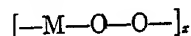
Oxygen, a well-known inhibitor of polymerization,^{59,59} yields a peroxide radical of rather low reactivity¹⁰⁴



but capable nevertheless of adding a monomer (styrene, methyl methacrylate, or vinyl acetate⁵⁹) to regenerate a normal chain radical with the ultimate production of polymer containing oxygen^{108,109}



Polymeric products having compositions approaching that for the 1:1 addition of oxygen to monomer, i.e.



are obtained.^{59,109} Barnes and co-workers⁵⁹ have confirmed this structure by hydrogenation and identification of the resulting glycol. The degrees of polymerization are low—about 10 to 40. Bovey and Kolthoff¹⁰⁹ found that the consumption of oxygen in the well-agitated emulsion polymerization of styrene was independent of the oxygen pressure. The rate depended directly on the initiator ($K_2S_2O_8$) concentration, however, and was about one-thousandth of the polymerization rate in the absence of oxygen. These facts, together with the occurrence of genuine induction periods in thermal or photochemical polymerizations conducted in the presence of limited quantities of oxygen, demonstrate that whereas reaction (60) supersedes monomer addition in speed, reaction (61) must be very slow indeed.

At the conclusion of the induction period due to oxygen, polymerization sets in at a rate exceeding that for pure monomer under the same conditions. The polymeric peroxides apparently furnish a source of free radicals. Oxygen therefore combines the roles of inhibitor, co-monomer, and (indirectly) of initiator.

The hazards of a rigid classification of substances which may modify the course of a free radical polymerization are well illustrated by the examples of inhibitors and retarders which have been cited. The distinction between an inhibitor or retarder, on the one hand, and a co-monomer or a transfer agent, on the other, is not sharply defined. Moreover, if the substance is a free radical, it is potentially either an initiator or an inhibitor, and it may perform both functions as in the case of triphenylmethyl. If the substance with which the chain radicals react is a molecule rather than a radical, three possibilities may arise: (i) The adduct radicals may be completely unreactive toward monomer. They must then disappear ultimately through mutual interaction, and we have a clear-cut case of either *inhibition* or *retarda-*